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TITLE: Low-Energy Positron-Matter Interactions Using Trap-Based Beams

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TITLE: Non-Neutral Plasma Physics 4. Workshop on Non-Neutral Plasmas
[2001] Held in San Diego, California on 30 July-2 August 2001

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Low-Energy Positron-Matter Interactions Using Trap-Based Beams

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Abstract. We present an overview of nonneutral plasma techniques developed to study the interaction of low-energy positrons with atoms and molecules. Both scattering and positron annihilation experiments are described. The scattering experiments provide the first state-resolved cross sections for both vibrational excitation of molecules and electronic excitation of atoms and molecules by positron impact. The annihilation experiments provide the first energy-resolved measurements of positron annihilation. Extensions of these techniques are briefly discussed, including work to create a new generation of positron beams with milli-electron volt energy resolution and the development of methods to study atomic clusters and dust grains.

I. INTRODUCTION

Positrons are now used routinely for a wide range of applications, including the study of atomic and molecular physics [1], plasma physics [2, 3], experiments attempting to form antihydrogen [4], and the characterization of materials and material surfaces [5, 6]. Nevertheless, progress in many of these areas has been limited by the availability of cold, intense positron sources. The situation has changed qualitatively by the recent exploitation of nonneutral plasma physics techniques to produce antimatter plasmas and beams in new regimes of parameter space and to manipulate these collections of positrons for specific measurements [2, 7, 8]. Methods were developed to efficiently accumulate and cool positrons from a ^{22}Na radioactive source. This source of cold positrons led to the development of a new method to create a cold, bright positron beam, tunable from 50 meV upwards, having an energy resolution of 20 meV, FWHM [9, 10]. These developments in positron plasmas and beams have enabled new studies of the interaction of low-energy positrons with ordinary matter. They have been used to study aspects of atomic and molecular physics and plasma physics, and they have sparked efforts in other areas, including a promising approach

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to the formation of neutral antihydrogen atoms [11] and new approaches to positron beam sources for the study of materials and material surfaces [12].

In this paper, we present an overview of recent studies of the interaction of positrons with atoms and molecules made possible by this technology. These advances have brought qualitative changes to the way in which positron scattering and annihilation experiments can be done. To put these developments into perspective, Fig. 1 shows the typical ranges of energies of important physical processes for positron interactions with atoms and molecules. The 20 meV resolution of the trap-based cold positron beam represents more than an order of magnitude improvement over conventional positron scattering experiments (e.g., with typical energy resolutions ≥ 400 meV). In the last two years, it has enabled the first measurements of cross sections for excitation of vibrational modes in molecules by positron impact [13, 14], the first state-resolved cross sections for electronic excitation [15], and the first energy resolved measurements of positron annihilation on atoms and molecules [16]. These results can be regarded as first steps in establishing a quantitative antimatter-matter chemistry, important not only in obtaining a fundamental understanding of nature, but also in using antimatter in the laboratory for science and technology.

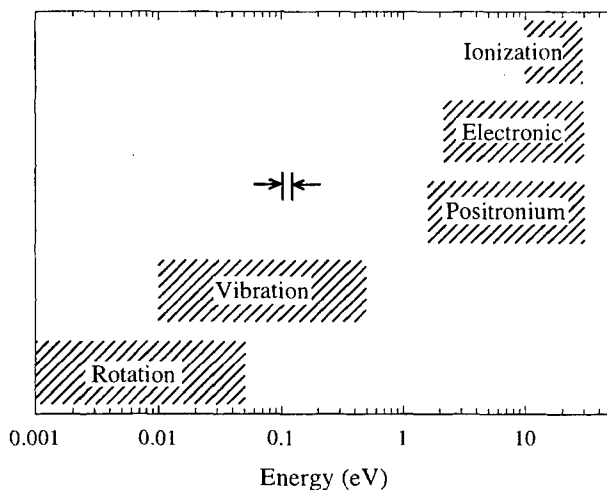


FIGURE 1. Representative energies of important processes in positron-matter interactions, including electronic excitation of atoms and molecules, vibrational and rotational excitation of molecules, positronium formation, and ionization. Annihilation (not shown) can occur at all energies. Vertical bars at 0.1 eV indicate the 20 meV energy resolution of the cold positron beam described here.

We describe here a new technique to make scattering measurements with the cold positron beam in the presence of an appreciable magnetic field (~ 0.1 tesla). This method is, in fact, superior to conventional positron and electron scattering techniques (e.g., using electrostatic beams) for a number of important measurements, such as energy-resolved cross sections for specific excitation processes integrated over scattering angle.

Annihilation is another important aspect of positron-matter interactions, and almost all positron annihilation studies to date have been done with a Maxwellian distribution at 300 kelvin. We describe very recent experiments that use the cold positron beam to make annihilation measurements as a function of positron energy. This technique, which is still in its infancy, provides important, target-specific information, such as the dependence of annihilation in molecules on the molecular vibrational modes.

In this paper we present an overview of these new methods to study positron scattering and annihilation processes -- techniques enabled by nonneutral plasma technology. We present examples of recent measurements made with these techniques and describe briefly future research directions. There are a wide range of problems in this area that can now be addressed, ranging from elucidating the mechanisms for positron annihilation in large molecules to understanding the fragmentation patterns of molecules following positron annihilation.

II. POSITRON ACCUMULATION AND COLD BEAM FORMATION

The buffer-gas trapping scheme is by far the most efficient of any method used to date to accumulate and cool large numbers of positrons [2, 17, 18]. Typically, $\sim 1\%$ of positrons from a ^{22}Na source are slowed to a few electron volts using a solid neon moderator. They are then injected into a Penning-Malmberg trap in the presence of a buffer gas and an applied magnetic field ~ 0.1 T. The accumulator has three stages, each at successively lower gas pressure and electrostatic potential. As many as 30% of the incident positrons become trapped in the third stage of the accumulator where they cool to room temperature in ~ 0.1 s [19, 20] on a mixture of N_2 and CF_4 . Using this technique, 3×10^8 e^+ have been accumulated in 8 minutes from a 90 mCi ^{22}Na source.

This trapping scheme was used to create a state-of-the-art cold positron beam (parallel energy spread ≤ 20 meV, FWHM) [9, 10], tunable over a wide range of energies, from ~ 0.05 eV upwards. This technique can be used to increase the brightness of a positron or electron beam and to create intense, short pulses of positrons with narrow energy spreads. In the case of positrons, it exceeds what has been done with other techniques in terms of energy resolution. In the case of electrons, while there are methods to generate much colder beams, this new method is likely to have advantages for specific applications such as study of the electron-beam positron-plasma instability [3, 21].

Cold beam formation is illustrated in Fig. 2. After the positrons are trapped and cooled, the potential of the bottom of the trap is raised, forcing the particles over a fixed-height potential barrier, V_B . The spread in parallel beam energies is measured using a retarding potential analyzer (RPA), by adjusting the voltage V_A in Fig. 2 (b). Using this technique, beams can be created with parallel energy spreads as small as 18 meV (FWHM). The perpendicular energy spread is comparable in magnitude (i.e. $\Delta E_{\perp} \sim k_B T = 25$ meV).

III. A NEW METHOD TO STUDY POSITRON SCATTERING

We have used the cold positron beam for a range of low-energy scattering studies [13, 14]. To do this, we developed a new technique to study scattering processes [22]. Since the positron plasmas and cold beam are in a magnetic field, it is convenient to also study the scattering in a magnetic field, in contrast to conventional atomic physics scattering experiments which are conducted with *electrostatic* beams in field-free collision environments. This new technique exploits the adiabatic properties of the positron's motion in the field -- a principle well known in the plasma community but not fully exploited for atomic physics applications. This technique is likely to have additional uses beyond the specific application to positrons (e.g., for electron scattering studies).

The arrangement for the scattering experiments is shown in Fig. 2. The positron beam is guided magnetically through a gas cell containing the test gas, then analyzed using a retarding potential analyzer (RPA) [15, 22]. The principle of the technique is illustrated in Fig. 3. It relies on the fact that the positron orbits are strongly magnetized (\sim e.g., for $B \sim 0.1$ T, the particle gyroradii are $\leq 10 \mu$). In this case, the total energy of the positrons can be separated into a sum of a component, E_{\parallel} , parallel to the magnetic field, and a second component, E_{\perp} , representing the energy in the particle gyromotion in the plane perpendicular to B . With the exception of the short duration of the scattering

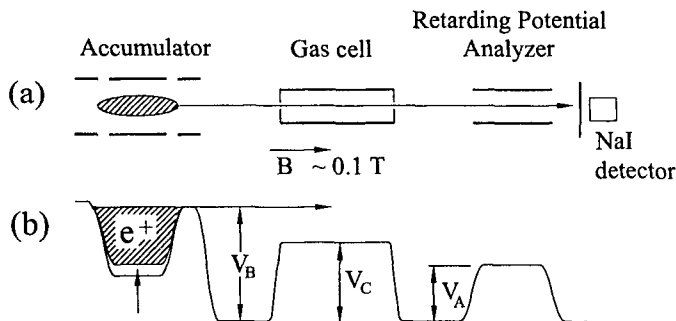


FIGURE 2. (a) Schematic diagram of the apparatus used to form a cold positron beam and to study scattering in a magnetic field; (b) the corresponding potential profile.

event, the quantity E_{\perp}/B is constant (i.e., an adiabatic invariant). The magnetic field in the scattering and analysis regions can be adjusted independently, and as we discuss below, this provides a convenient way of manipulating the perpendicular and parallel energy components of the positrons for specific measurements.

To date the positron beam and this scattering technique have been used for studies of total cross sections (TCS), differential elastic cross sections, and 'integral cross sections' (i.e., cross sections for *specific inelastic processes* integrated over angle). Here we present examples of the first measurements ever taken of integral cross

sections for both electronic and vibrational excitation of molecules by positrons [13-15]. Figure 4 shows data for a measurement of the vibrational excitation cross section of CO at 0.5 eV [13]. In Fig. 4(a) is shown an RPA curve when the ratio of fields, M , in the scattering and analysis region are the same. This corresponds to the situation illustrated in Fig 3(a) in which the RPA measurement is unable to distinguish the elastic and inelastic scattering components. In contrast, data taken at a field ratio $M = 35$ are shown in Fig. 4(b). This situation corresponds to that illustrated in Fig. 3(b). There is now a distinct separation of the inelastic and elastic scattering. This results in a sharp step at an RPA voltage corresponding to the *total* energy of a positron after an inelastic scattering event, while the elastic scattering is compressed in a small energy region near the beam cut-off. Note the expanded scale in (b), the good sensitivity, and the excellent discrimination of the inelastic process.

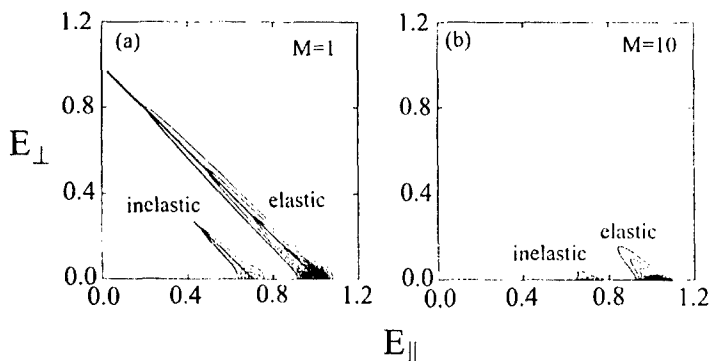


FIGURE 3. Simulation of the scattering of particles from a beam with energy $E_{\parallel} = 1.0 \gg E_{\perp}$, initially traveling parallel to the magnetic field. (a) Elastic and inelastic scattering. (b) For a magnetic field ratio of $M = 10$ between the scattering cell and the RPA, the spread in E_{\perp} is greatly reduced, so an RPA measurement can distinguish elastic and inelastic scattering.

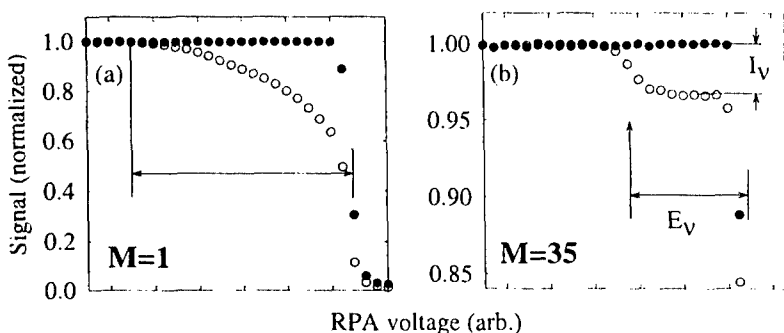


FIGURE 4. RPA measurement for CO using a 0.5 eV positron beam (open circles); no gas in cell (solid circles): (a) magnetic field ratio, $M = 1$, of the field in the scattering cell to that in the analysis region; (b) same measurement at $M = 35$. The step marked by the arrow in (b) corresponds to excitation of the vibrational mode in CO at $E_v = 0.27$ eV.

Figure 5 shows examples of measurements for vibrational excitation of molecules by positrons. Data for H_2 and CO_2 are shown together with available theoretical predictions [13]. Note that two modes are resolved in CO_2 , with the lowest having an energy of only 0.08 eV. Comparisons with theory are absolute, with no fitted parameters. Agreement with theory is very good considering this is the first time such a comparison was possible. Work is currently in progress to examine in more detail the behavior of the cross sections near threshold.

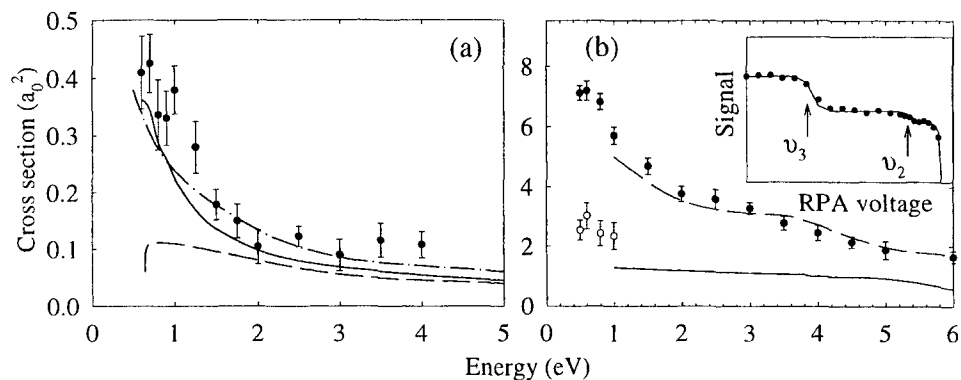


FIGURE 5. Cross sections for vibrational excitation as a function of positron energy: (a) the v_1 mode of H_2 at 540 meV, compared with theoretical predictions: solid line, Ref. [23]; dot-dashed line, Ref. [24]; and dashed line, Ref. [25]. (b) the v_2 and v_3 modes in CO_2 at 80 and 290 meV, respectively. The inset in (b) shows raw data and resolution of the 80 meV, v_2 excitation. The lines (except in the inset), are comparisons with the theory of Ref. [26].

Figure 6 shows examples of the first state-resolved cross sections for *electronic* excitation of molecules [15]. These measurements required resolving structure due to the vibrational manifolds of the excited electronic states; which was made possible because of the excellent energy resolution of the cold beam. The reader is referred to Ref. [15] for details.

To put these measurements in perspective, also shown in the figure are the only measurements of cross sections for excitation of these transitions by electron impact. Comparison of the two data sets indicates that the quality of the positron data are comparable to or better than the electron measurements. This is due to the fact that the electron measurements were done with an electrostatic beam, measuring the differential cross section at many angles and then integrating over angle. This procedure is relatively time consuming and error prone for this kind of measurement, as compared with the technique described here.

The data for H_2 strongly favor one of two competing theoretical calculations. There are no theoretical predictions presently available for N_2 . The sharp rise in the cross section in N_2 near threshold provides insight into the operation of the buffer-gas

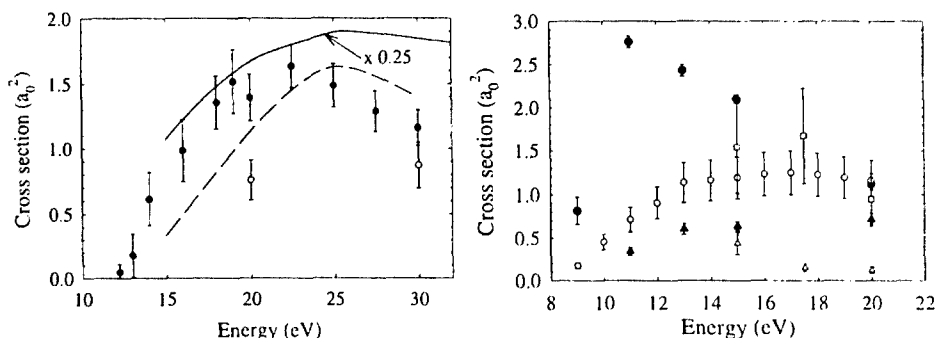


FIGURE 6. (a) Cross section for excitation of the $B^1\Sigma$ electronic state of H_2 , and absolute comparison with theory: solid line, Ref. [27] and dashed line, Ref. [28]. (b) Cross section for excitation of the $a^1\Pi$ and $a^1\Sigma$ states of N_2 are shown by the solid circles and triangles respectively. Open symbols are the available electron scattering data: (a) circles, Ref. [29]; (b) circles, Ref. [30], and squares and triangles, Ref. [31].

positron trap. Empirically, the buffer-gas trap works most efficiently on N_2 when the energy loss per stage is tuned to operate in this region (i.e., $\sim 9 - 10$ eV). In N_2 , positronium formation (which is the dominant loss process in the trap), begins at a threshold energy of 8.8 eV, increasing from zero at that point. The data in Fig. 6(b) indicate that the high trapping efficiency in N_2 is due to the very rapid increase in the cross section for electronic excitation in the region just above threshold where the loss due to positronium formation is relatively small.

IV. ENERGY RESOLVED ANNIHILATION MEASUREMENTS

The fate of all antimatter in our world is annihilation with ordinary matter. Thus understanding the details of these annihilation processes is both of fundamental importance and crucial for a range of applications. With the exception of experiments conducted in the buffer-gas trap varying the temperature of the positrons, studies of positron annihilation have been limited to room temperature. An outstanding problem for almost four decades is that positron annihilation on molecules can be orders of magnitude larger than expected from simple collisions. In order to address this problem, we recently began experiments using the cold beam to make high-resolution measurements of annihilation as a function of positron energy [16]. These measurements, which are the first of their kind, are difficult due to the fact that the annihilation cross section is typically several orders of magnitude smaller than the cross sections for typical scattering processes (i.e., those illustrated in Fig. 1).

The apparatus for the annihilation experiment is shown schematically in Fig. 7. In this experiment, $2 \mu s$ pulses of $\sim 5 \times 10^4$ cold positrons are passed through the annihilation cell. Annihilation is monitored from a 10 cm long region along the positron beam. In order to minimize the background signal that would arise if the

positrons were to strike a collector or other metal surface, the positrons are kept in flight, transiting the cell ~ 10 times during the time annihilation gamma rays are monitored. The detector consists of a relatively compact CsI crystal with integrated light pipe and photodiode surrounded by a lead shield. Typical background levels are one count per 10^9 positrons.

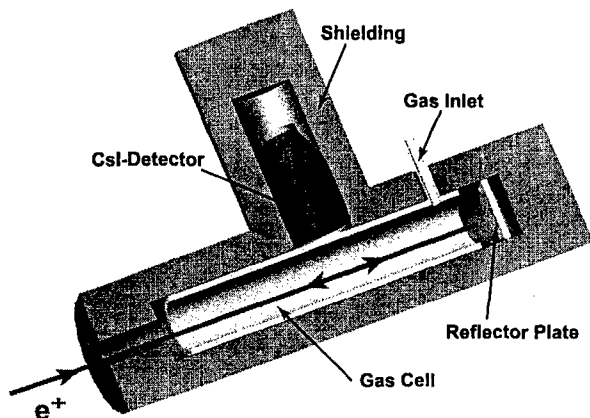


FIGURE 7. The apparatus used for energy-resolved positron annihilation measurements. Monoenergetic pulses of positrons transit the gas cell while the gamma ray signal is recorded.

Thus far we have studied annihilation from Xe atoms and several molecules (mostly hydrocarbons) ranging in size from acetylene (C_2H_2) to butane (C_4H_{10}) in the range of energies from 50 meV to ~ 500 meV. Selected studies have included higher energies, up to ~ 4 eV.

Shown in Fig. 8 are data for annihilation in butane from an energy of 50 meV to within 0.5 eV of the positronium threshold at 3.8 eV [16]. The annihilation rate is given in terms of the dimensionless parameter, Z_{eff} , defined in terms of the Dirac annihilation rate for a positron in an electron gas, assuming no correlation of the particles. If this simple model were correct, the annihilation rate would be $Z_{eff} = Z$, where Z is the number of electrons in the molecule.

The data for butane are typical of the alkane molecules studied (C_2H_6 , C_3H_8 , and C_4H_{10}), in that very large enhancements in the annihilation rate are observed when the incident positron energy is tuned to the range of the molecular vibrations, while Z_{eff} is ≤ 100 at energies above this region. In butane, the sharp peak observed at the C-H stretch mode (i.e., at ~ 0.36 eV) is 23,000, compared with the value of 10,500 measured with a Maxwellian distribution of positrons at 300 K.

While these experiments are in their infancy, a number of important results have already emerged. The data show clearly that it is vibrational resonances of the positron molecule complexes that lead to the large values of Z_{eff} . The resonances in larger molecules are downshifted tens of millivolts; this is interpreted as a measure of

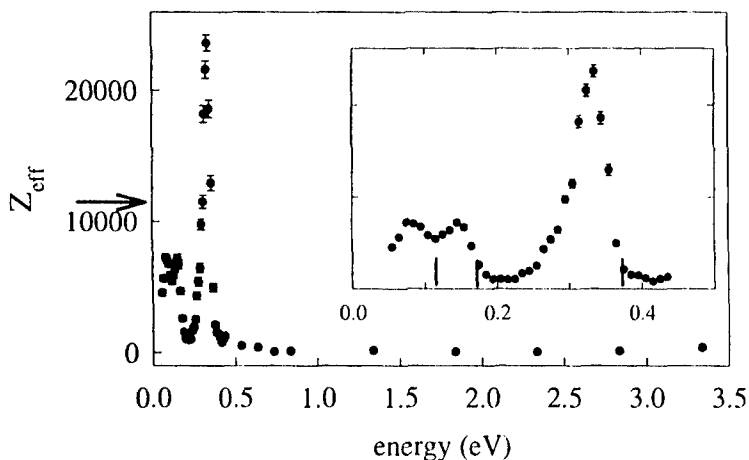


FIGURE 8. The first energy resolved measurements of positron annihilation rates. The normalized rate, Z_{eff} , is shown for butane (C_4H_{10}) as a function of positron energy, ϵ , in the range $50 \text{ meV} \leq \epsilon \leq 3.3 \text{ eV}$. The inset shows the same data in the range, $50 \leq \epsilon \leq 450 \text{ meV}$, on an expanded scale. The arrow on the ordinate indicates the value at room temperature. The vertical bars on the abscissa in the inset indicate the positions of the infrared-active vibrational modes.

the binding of positrons to molecules [16]. Finally, addition of fluorine atoms to hydrocarbons results in a decrease in Z_{eff} at the C-H stretch mode energy by a much larger factor than the change in the fraction of C-H bonds in the molecule. This tends to support the conjecture of Gribakin [32, 33] that fluorination of hydrocarbons can change the positron-molecule potential from attractive to repulsive, and turn off the resonant annihilation mechanism.

V. A LOOK TO THE FUTURE

There are several new experiments made possible by the positron scattering and annihilation techniques described here, and a number of potentially important extensions of these techniques are now possible. Experiments are currently in progress to study several aspects of positron scattering. We are interested in understanding the sharp threshold behavior in vibrational excitation of molecules, more complete measurements in energy and more detailed comparisons with theoretical calculations. The subject of electronic excitation by positron impact is even less mature, and our measurements have now motivated new theoretical efforts [34]. Questions of interest include obtaining a quantitative understanding of the cross sections and effects such as the threshold behavior of the $\text{A}^1\Pi$ excitation cross section in N_2 (c.f., Fig. 6). Another question of interest is whether positron impact can result in electronic excitations of targets involving a spin flip. Such transitions, which are common in electron scattering due to exchange, might occur via the spin-orbit interaction in the case of positrons. We have also searched briefly for resonances in

scattering in the range of energies of low-lying electronic excitations. Further studies of this phenomenon are planned.

There are a large number of open questions in the area of positron annihilation that can now be addressed by the new experiment described above. Our first experiments appear to confirm the model of Gribakin that large values of annihilation in molecules occurs *via* vibrational resonances of the positron-molecule complex. One can now make more detailed tests of the model. Questions of interest include the magnitude of the positron binding energy, the density of resonances as a function of energy, and the dependence of these resonances on molecular structure.

Several extensions of the work in a number of directions are now either in progress or are planned. As indicated in Fig. 1, good energy resolution is crucial in studying many important processes. We are currently building a positron trap having a 5 tesla magnetic field and walls cooled to 10 kelvin. The plasma will cool to 10 kelvin (equivalent to 1 meV) by cyclotron radiation. This device should, in principle, be capable of producing a positron beam with millielectron volt energy resolution which would enable a new regime of high resolution scattering and annihilation studies.

Other planned directions include study of the scattering and annihilation of positrons interacting with larger molecules (e.g., those with negligible vapor pressure at room temperature), atomic clusters and dust grains. Neutral large molecules and clusters can be studied in a gas cell at elevated temperature. Ionized species and ionized fragments produced *via* annihilation can be studied *in situ* in a Penning trap in the high field magnet.

VI. CONCLUDING REMARKS

The understanding of antimatter-matter interactions has lagged significantly compared with analogous studies involving ordinary particles. In the past few years, progress has been made in closing this gap by research efforts such as that described here, research that was driven in large part by novel applications of nonneutral plasma techniques. Many important questions in this area are either presently being studied or can now be addressed. Additional tools for these studies, such as a millielectron volt positron beam, are currently under development. This progress bodes well for achieving a fundamental understanding of broad classes of antimatter-matter interactions that can be used, in turn, for a range of scientific and technological applications.

ACKNOWLEDGEMENTS

We are pleased to acknowledge helpful conversations with Rod Greaves, Gleb Gribakin, and Jim Mitroy, and the technical assistance of Gene Jerzewski. This work is supported by the National Science Foundation, grant PHY 98-76894, and the Office of Naval Research, grant N00014-97-1-0366. One of us (SJB) gratefully acknowledges the support of the Australian-American Fulbright Commission.

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